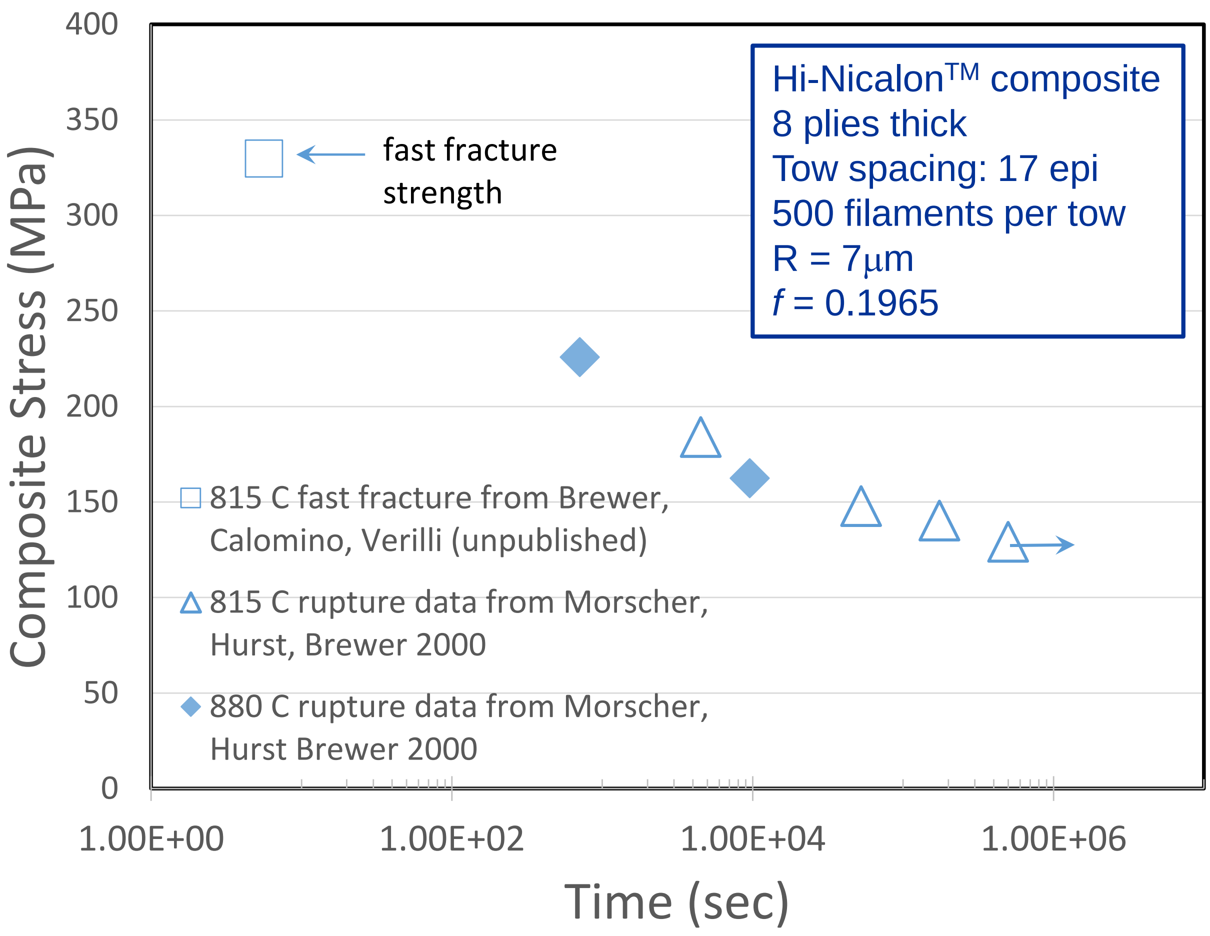
A Numerical Solution Routine for Investigating Oxidation-Induced Strength Degradation Mechanisms in SiC/SiC Composites

Roy M. Sullivan
Materials and Structures Division
NASA Glenn Research Center
Cleveland, OH

MS&T15
MATERIALS SCIENCE & TECHNOLOGY

Materials Science & Technology 2015
October 4-8, 2015
Greater Columbus Convention Center
Columbus, OH, USA

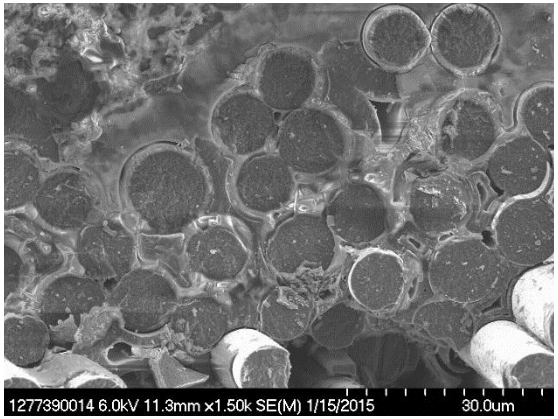
What causes the time dependent strength degradation in SiC/SiC composites at intermediate temperatures (700 – 900 °C)?



Stress versus time-to-failure of Hi-Nicalon™ composite specimens at intermediate temperatures from Morscher, Hurst and Brewer (2000).

Time Dependent Strength Degradation Mechanisms

Theory #1: Oxidation of BN fiber coating causes fusing of fibers to one another and to matrix resulting in embrittled composite.



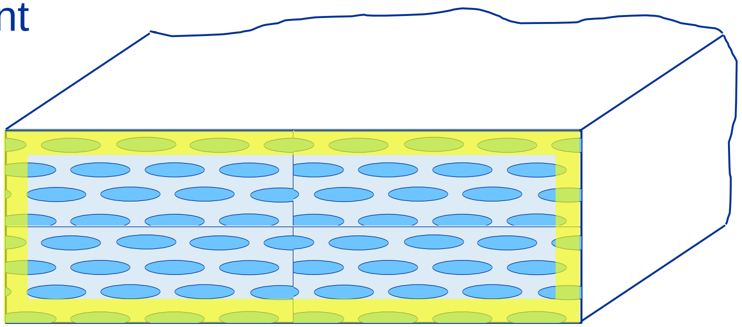
Micrograph of SiC/SiC composite showing oxidized BN fiber coating. Courtesy of Ram Bhatt (NASA/GRC).

Heredia, et al. (1995)
Morscher (1997)
Glime and Cawley (1998)
Morscher, Hurst and Brewer (2000)
Morscher and Cawley (2002).

$$2BN + \frac{3}{2}O_2 \rightarrow B_2O_3 + N_2$$

$$B_2O_3 + SiO_2 \rightarrow borosilicate\ glass$$

- Fusing causes local load sharing (LLS): Fibers adjacent to failed fibers are overloaded, causing a cascading of fiber failures and composite failure.
- Embrittlement is time dependent since extent of the cross-section that is fused increases with time.

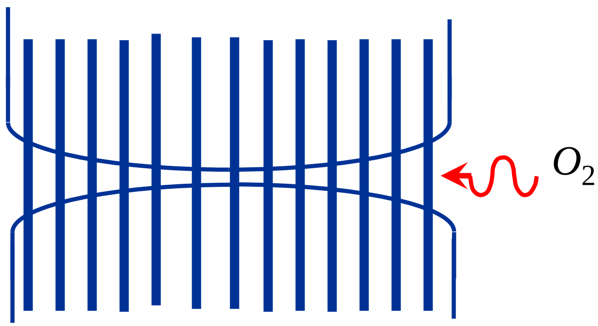


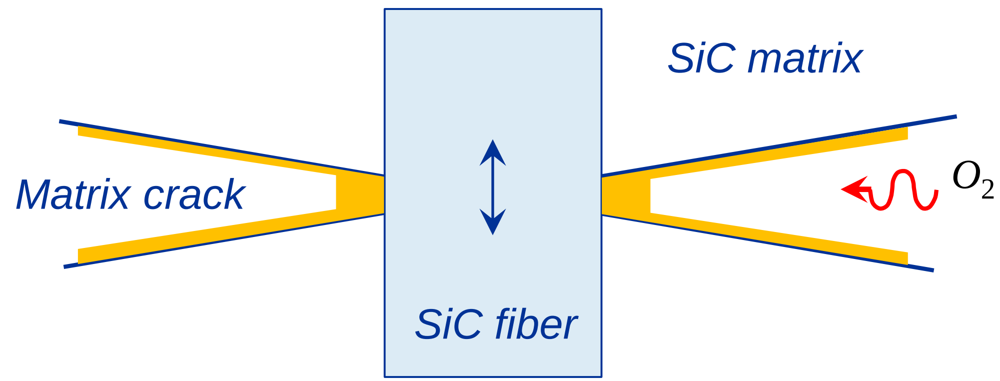
Time Dependent Strength Degradation Mechanisms

Theory #2: Oxidation of SiC fiber results in tensile stress in fiber.

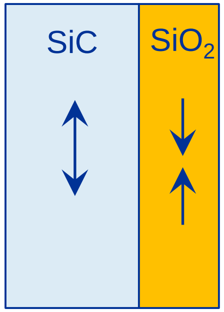
Xu, Zok and McMeeking (2014)
Hay (2012).

$$SiC + 2O_2 \rightarrow SiO_2 + CO_2$$





- Molar volume of silica is greater than SiC causing compression in oxide and tension in fiber.
- Tensile stress in fiber increases with time since oxide thickness grows with time.
- Results in an apparent loss in fiber strength over time.



Fiber with oxide scale

Time Dependent Strength Degradation Mechanisms

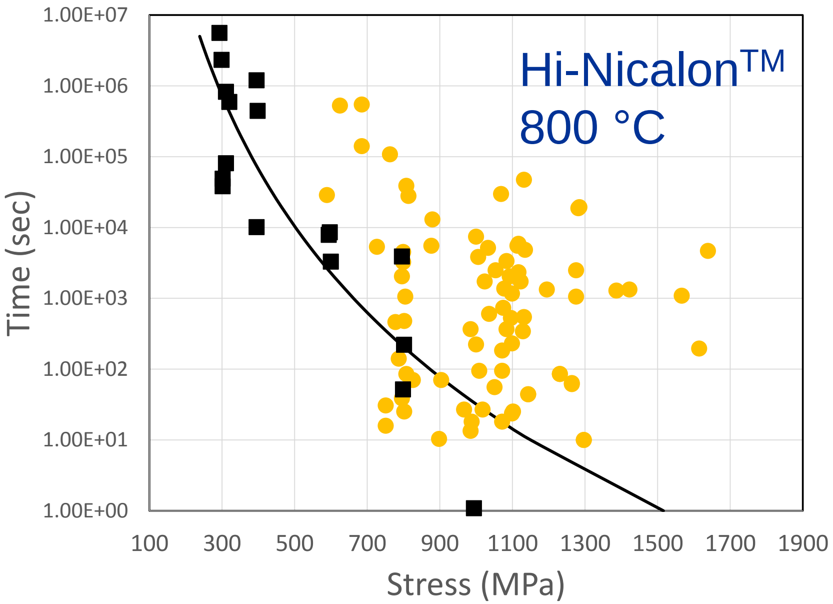
Theory #3: SiC fiber strength is intrinsically time dependent due to slow crack growth in fibers.

Forio, Lavaire and Lamon (2004)
Gauthier, Pailler, Lamon and Pailler (2009)
Gauthier and Lamon (2009).

At intermediate temperatures, tows and single fibers have a stress vs time-to-failure relationship that follows

$$\sigma_f^n\, t = A$$

Some evidence of slow crack growth on fiber fracture surfaces



Rupture time versus stress for Hi-Nicalon™ single filaments and tows at 800 °C. Data from Gauthier and Lamon (2009).

Objective

- Investigate the cause of the time-to-failure vs. stress relationship in SiC/SiC composites with a BN fiber coating at intermediate temperatures.

Approach

- Develop a progressive failure analysis routine (based on Theory #3) and apply it to simulate the composite stress rupture tests that produced the results shown on the first slide. The ability to simulate the stress vs. time-to-failure behavior will judge its validity.

Assumptions

- Composite failure initiates at a matrix crack.
- The progression of fiber failure occurs under global load sharing (GLS).

Fiber Failure Model (Relationship between $P_f - \sigma - t$)

Fast Fracture Strength

$$C_i = \left(\frac{K_{Ic}}{\sigma_f^s Y} \right)^2$$

Flaw Growth Velocity (Davidge, et al., 1973)

$$v = \frac{dC}{dt} = \alpha_1 K_I^n$$

Weibull FF Strength Distr.

$$P_f = 1 - \exp\left(-\frac{L}{L_o} \left(\frac{\sigma_f}{S_o} \right)^m \right)$$

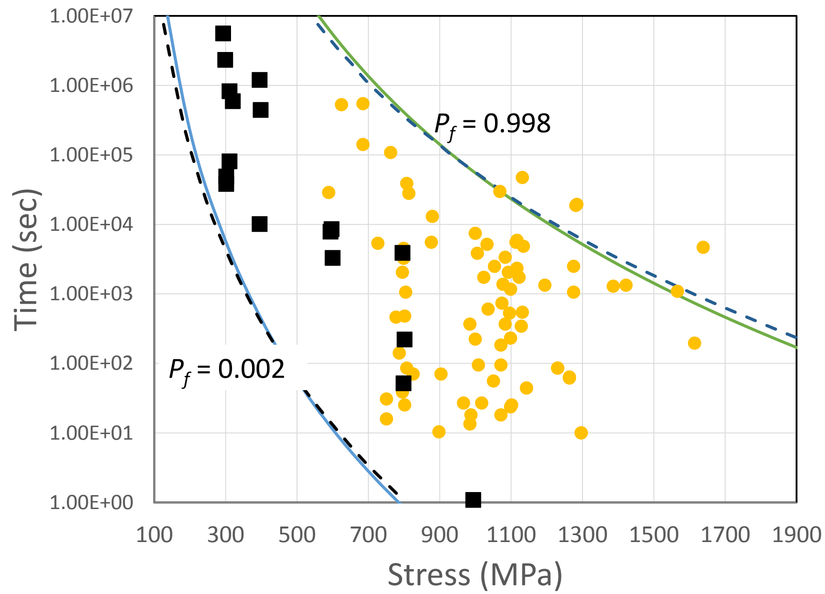
$$\sigma_f^n t = \frac{2}{\alpha_1 Y^2 (n-2)} \left(\frac{S_o}{K_{Ic}} \right)^{n-2} \left(\frac{L_o}{L} \ln \frac{1}{1-P_f} \right)^{\frac{n-2}{m}}$$

Fitting the probability of failure curves to the data, one can extract values for the constants

n α_1 *Flaw Growth Parameters*

m S_o *Weibull Statistical Parameters*

K_{Ic} *Fracture Toughness*



Time-to-failure versus applied stress for Hi-Nicalon™ single fibers (gold circles) and tows (black squares) at 800 °C from Gauthier and Lamon (2009).

Rearrange the previous expression to get an expression for the Probability of Failure

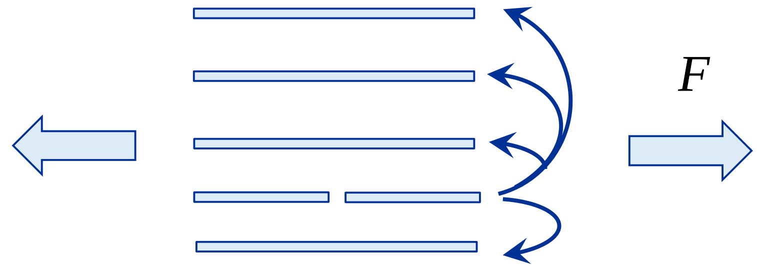
$$\sigma_f^n t = \frac{2}{\alpha_1 Y^2 (n-2)} \left(\frac{S_o}{K_{Ic}} \right)^{n-2} \left(\frac{L_o}{L} \ln \frac{1}{1-P_f} \right)^{\frac{n-2}{m}}$$

$$P_f = 1 - \exp\left\{ -\frac{L}{L_o} \left(\frac{1}{2} Y^2 \alpha_1 (n-2) t \right)^{\frac{m}{n-2}} \left(\frac{K_{IC}}{S_o} \right)^m \sigma_f^{\frac{nm}{n-2}} \right\}$$

Length effect

Progressive Failure Analysis (PFA) Routine

- Numerically similar to Lara Curzio (1997)
- Based on Global Load Sharing (GLS) Model



Global Load Sharing (GLS) Model

Flowchart

- Add time step, $t^{i} = t^{i-1} + \Delta t$

- Iterate between two equations

Prob. of Survival

$$1 - P_f = \frac{N}{N_o} = \exp\left[-\frac{L}{L_o} \left(\frac{1}{2} Y^2 \alpha_1 (n-2) t \right)^{\frac{m}{n-2}} \left(\frac{K_{IC}}{S_o} \right)^{m} \sigma_f^{\frac{nm}{n-2}} \right]$$

no

N

Force Equilibrium

$$\sigma_f = \frac{N_o}{N} \frac{F}{N_o A_f}$$

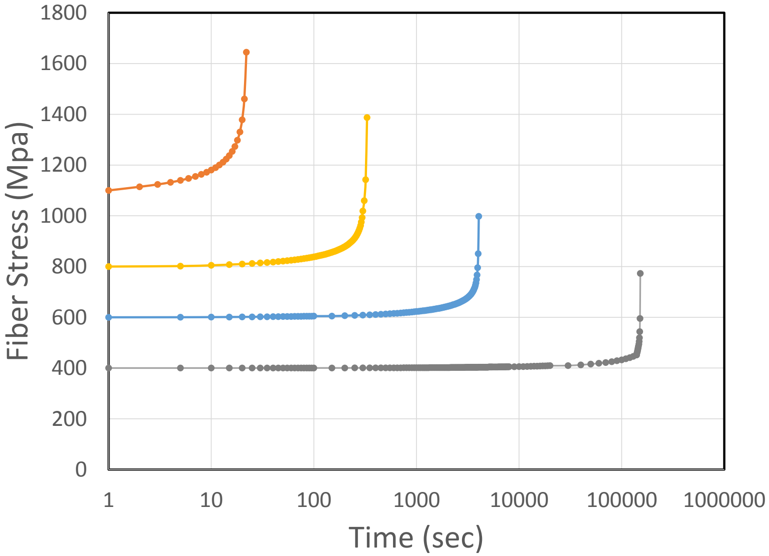
σ_f

Converge?

yes

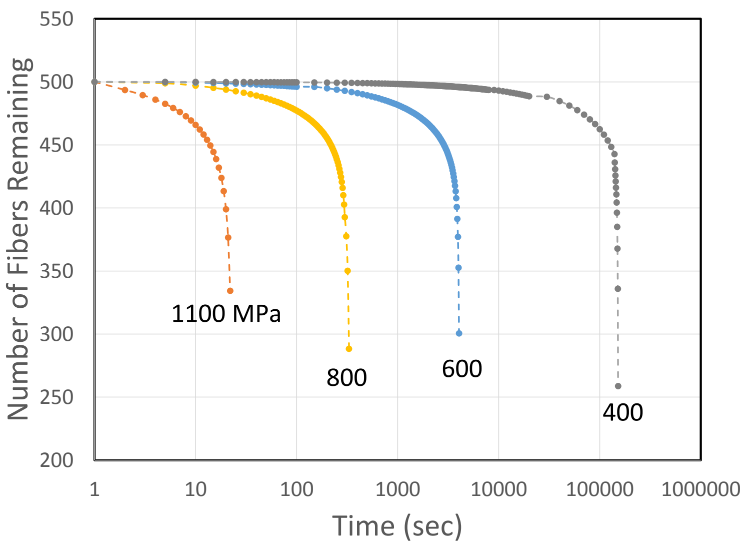
N_o	*number of fibers initially*
N	*number of fibers that are not failed*
A_f	*Cross-sectional area of fibers*

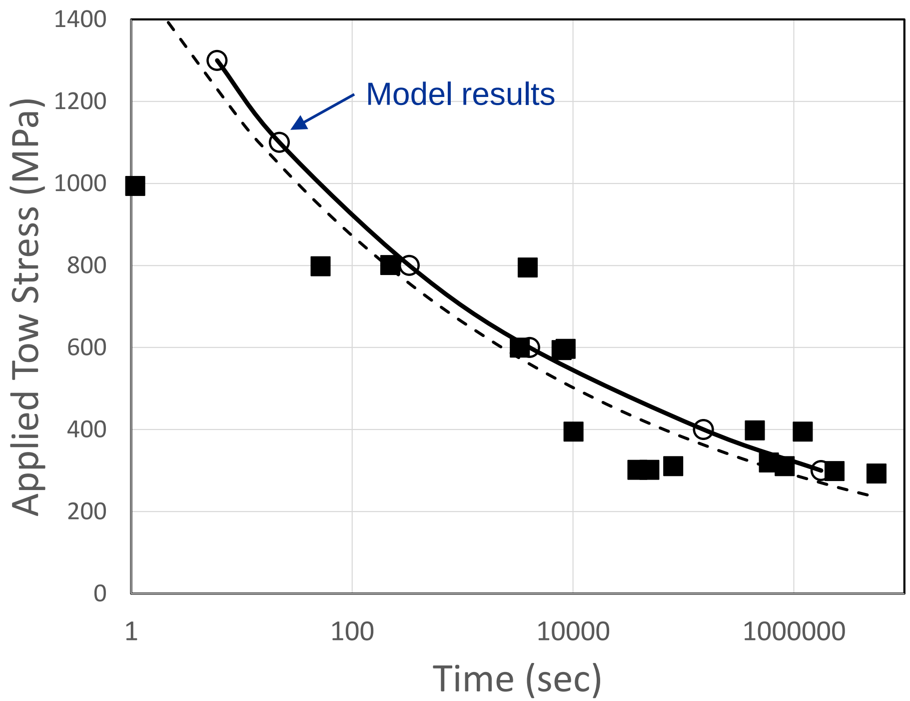
PFA Simulation of Tow Tensile Tests from Gauthier and Lamon (2009)



$$\sigma_f = \frac{F}{NA_f}$$

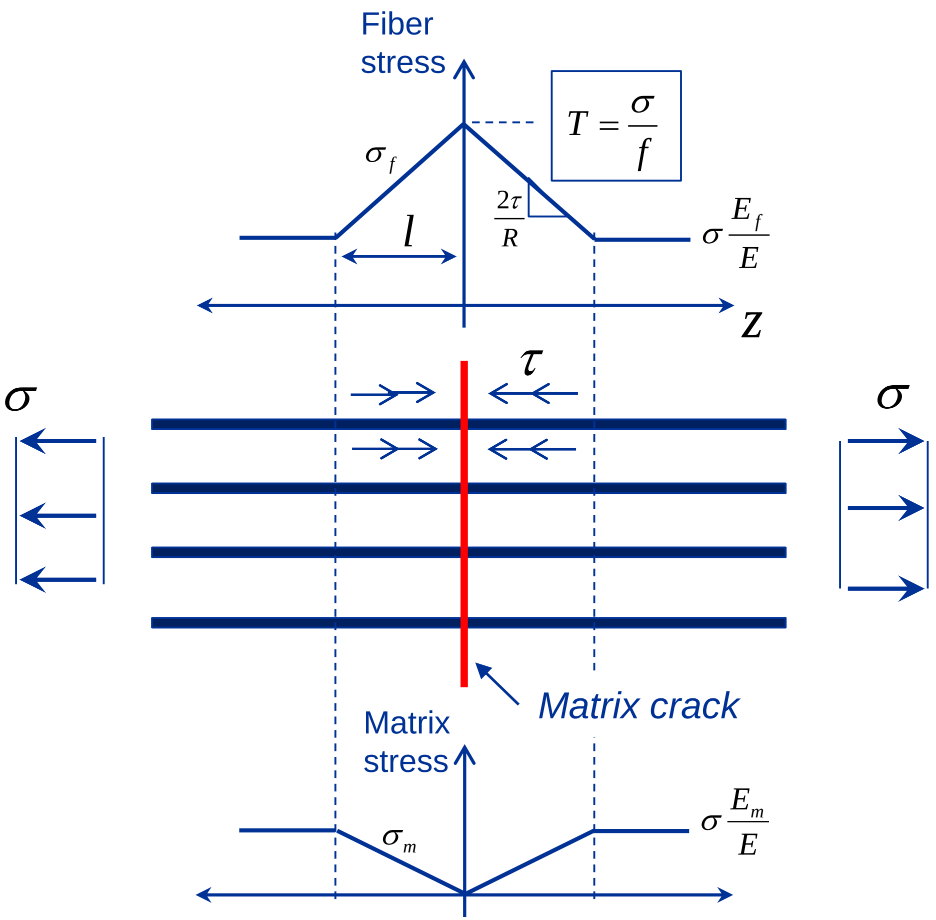
$$N_o = 500$$
$$L_o = 25\,mm$$





Analysis of Composite Systems

Assumption: Composite failure initiates at a matrix crack.



Aveston, Cooper and Kelly (1971)
Curtin (1991)
Curtin (1994)
Curtin, Ahn and Takeda (1998)
Thouless and Evans (1988)

Slip length

$$l = \frac{R}{2\tau}\left(\frac{\sigma}{f}\right)\frac{(1-f)E_m}{E}$$

R the fiber radius

f the fiber volume fraction in loading direction

τ the sliding resistance shear stress

Analysis of Composite Systems

Length of fiber loading is now 2*l* and probability of failure now involves fiber stress at crack plane *T*

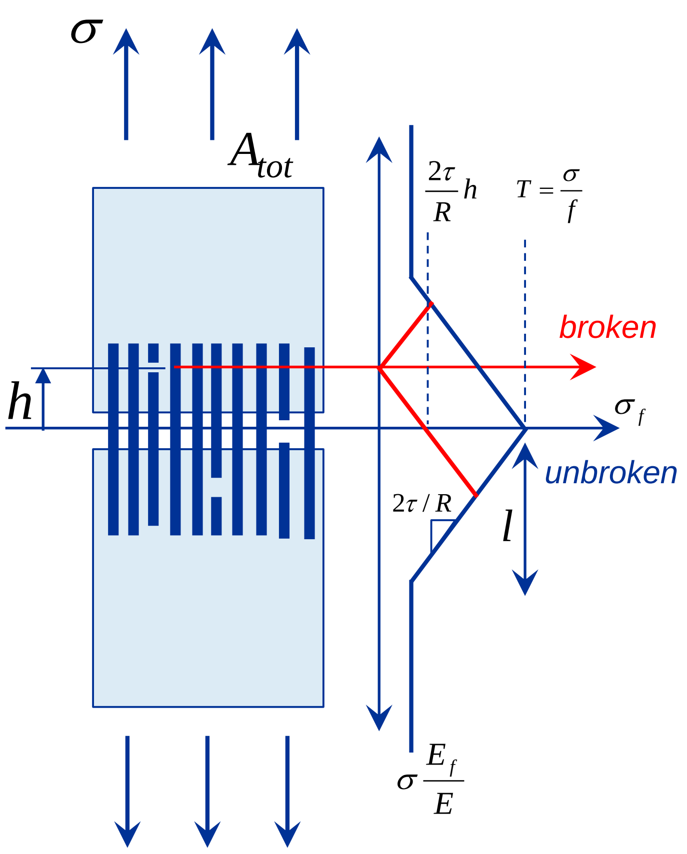
$$P_f = 1 - \exp\left\{ -\frac{2l}{L_o} \left(\frac{1}{2} Y^2 \alpha_1 (n-2) t \right)^{\frac{m}{n-2}} \left(\frac{K_{IC}}{S_o} \right)^m \frac{T^{\frac{mn}{n-2}}}{\left(\frac{mn}{n-2} + 1 \right)} \right\}$$

$$l = \frac{R}{2\tau} \left(\frac{\sigma}{f} \right) \frac{(1-f)E_m}{E}$$

Analysis of Composite Systems

For a single matrix crack, force equilibrium at crack plane requires

$$F = \sigma A_{tot} = \underbrace{TNA_f}_{\text{Force carried by unbroken fibers}} + \underbrace{(N_o - N)\langle\sigma_p\rangle A_f}_{\text{Force carried by broken fibers via pullout stress}}$$



$$\langle\sigma_p\rangle = \frac{2\tau}{R}\langle h\rangle$$ *Average Pull-out stress*

$\langle h\rangle$ *Average Pull-out length**

Rearranging

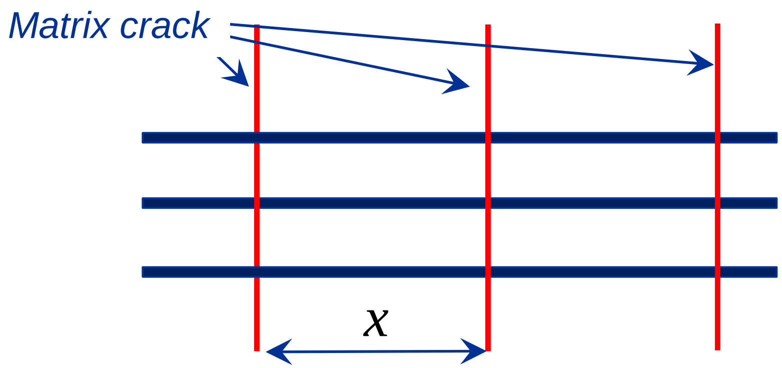
$$T = \frac{N_o}{N}\left[\frac{F}{N_o A_f} - \left(1 - \frac{N}{N_o}\right)\langle\sigma_p\rangle\right]$$

** Expression for average pull-out length obtained from Thouless and Evans (1988)*

Crack Spacing and Shear Stress Calculations

Shear stress can be estimated from crack density measurements when cracks are saturated.

Aveston, et al. (1971)
Kimber and Keer (1982)
Curtin (1991)

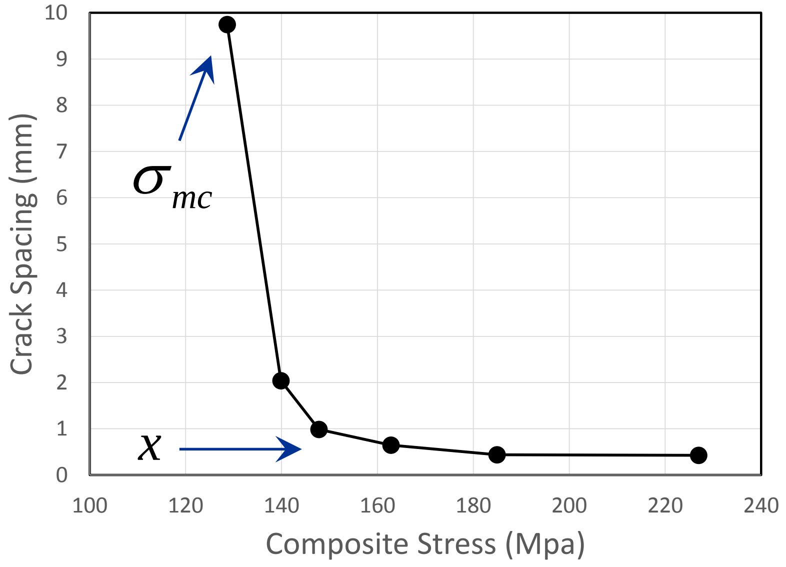


$$\bar{x} = 1.337 \frac{R}{2\tau}\left(\frac{\sigma_{mc}}{f}\right)\frac{(1-f)E_m}{E}$$

$$x = 0.4\,mm \qquad R = 7\,\mu m$$

$$\sigma_{mc} = 130 MPa \qquad f = 0.1965$$

$$\tau \approx 5\,MPa$$

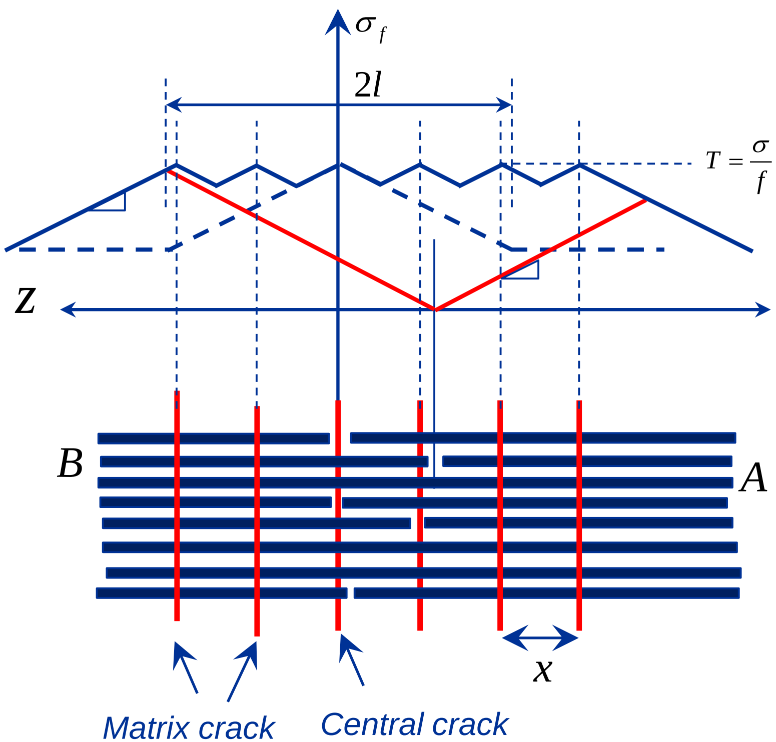


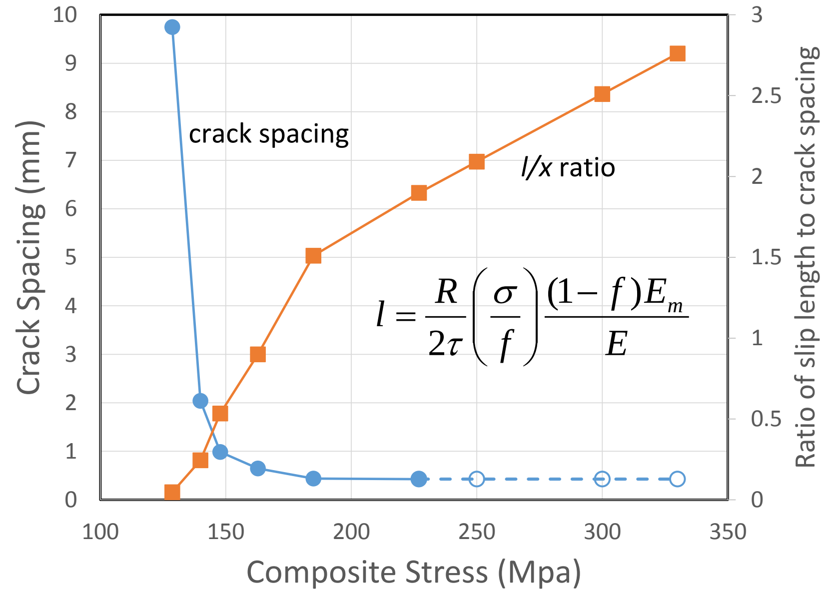
Crack density versus composite stress for Hi-Nicalon™ composite from Morscher and Cawley (2002)

Multiple Matrix Cracks within the Slip Length of One Another

If matrix cracks are close enough, fiber failures in nearby matrix cracks affect the force equilibrium equation in a central matrix crack plane

Number of matrix cracks within the slip length of any one matrix crack $\left(1+\frac{2l}{x}\right)$





Crack spacing and the ratio l/x versus composite stress. Crack spacing from crack density data from Morscher and Cawley (2002).

Force Equilibrium at Central Crack Plane (Curtin, et al. (1998))

$$T=\frac{F}{N_oA_f\left[1-\left(1-\frac{N}{N_o}\right)\left(1+\frac{l}{x}\right)\right]}$$

Analysis of Composite Systems

Progressive Failure Analysis routine now involves:

Prob. of survival

$$1-P_f = \frac{N}{N_o} = \exp\left\{-\frac{2l}{L_o}\left(\frac{1}{2}Y^2\alpha_1(n-2)t\right)^{\frac{m}{n-2}}\left(\frac{K_{IC}}{S_o}\right)^m \frac{T^{\frac{mn}{n-2}}}{\left(\frac{mn}{n-2}+1\right)}\right\}$$

Iteration *Iteration*

Force equilibrium

$$T = \frac{N_o}{N}\left[\frac{F}{N_o A_f} - \left(1-\frac{N}{N_o}\right)\langle\sigma_p\rangle\right]$$

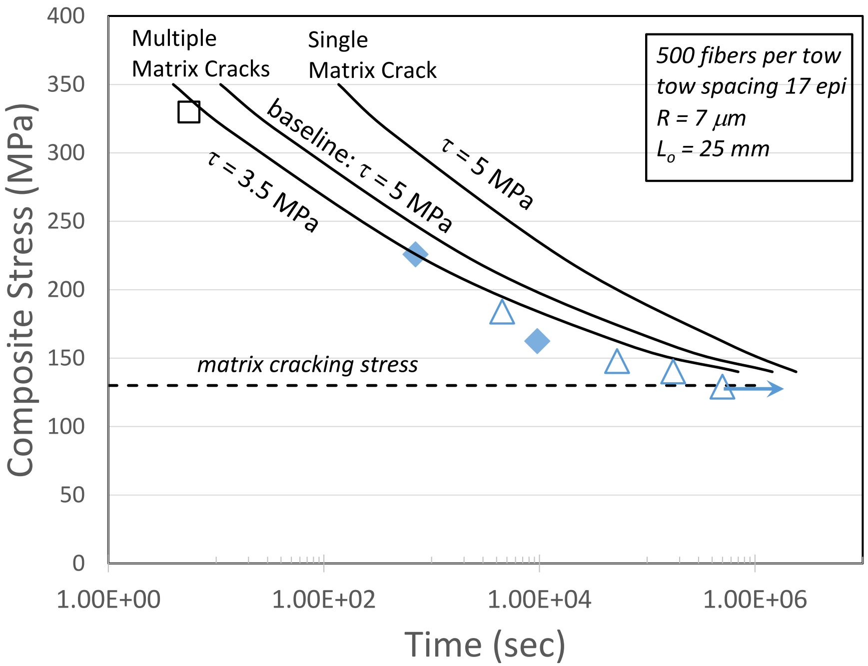
$$T = \frac{F}{N_o A_f\left[1-\left(1-\frac{N}{N_o}\right)\left(1+\frac{l}{x}\right)\right]}$$

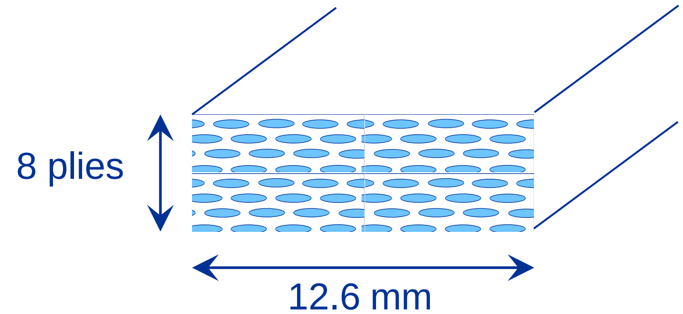
Single Matrix Cracks

Multiple Matrix Cracks

Analysis of Composite Systems

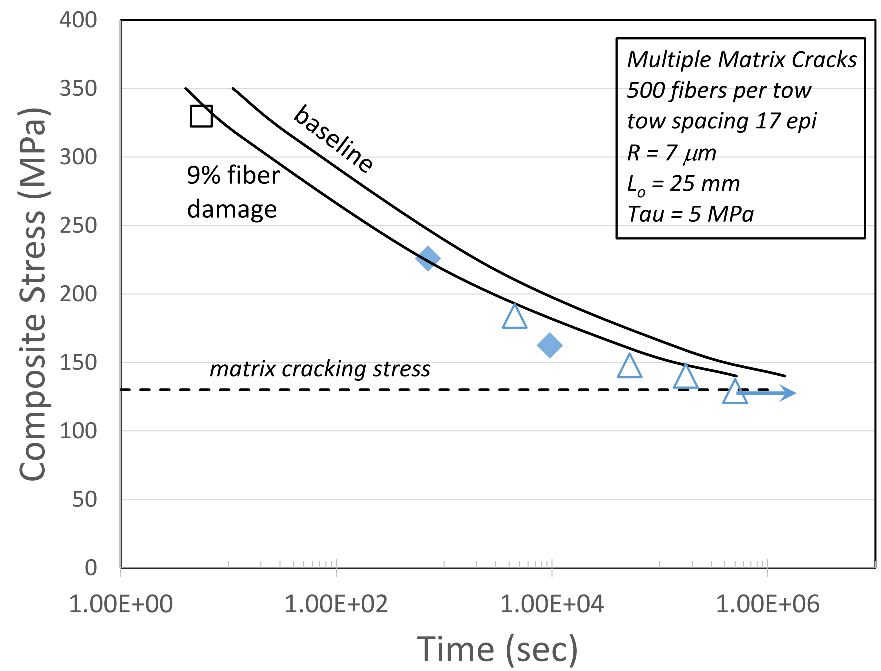
Results: Each line represents a series of PFA solutions





$$\left(\frac{12.6mm}{25.4}\right)(17epi)(8\,plies) \approx 67.5\ tows$$

$$N_o = 67.5(500) = 33{,}750$$



Note: Marshall and Evans (1985) measured a shear stress value in other SiC fiber-reinforced ceramic matrix composites in the range of 2 - 2.5 MPa.

Discussion and Conclusions

- The time dependent strength of Hi-Nicalon™ fiber reinforced composites has been shown to be largely due to the intrinsic time dependent strength of the fibers. Other mechanisms (e.g. fusing and embrittlement) may have a small effect at later times.

- Best agreement with the measured time-to-failure versus composite stress was obtained with progressive failure analyses solutions using multiple matrix crack formulation and with a combination of shear stress values between 3.5 – 5 MPa and fiber damage values of < 9%.

- If slow crack growth in fibers requires oxidation of inter-granular interface, what is the source of oxygen? Does it flow from the surrounding atmosphere down a matrix crack or is there enough present in the constituents? SiC fibers? BN fiber coating?

List of References

Aveston J., Cooper G.A. and Kelly A., “Single and Multiple Fracture,” pp. 15-26 in the Properties of Fiber Composites, Conference Proceedings of the National Physical Laboratory. IPC Science and Technology Press Ltd. Surrey, England, 1971.

Curtin W.A., “Theory of Mechanical Properties of Ceramic Matrix Composites,” *J. Am. Ceram. Soc*., **74** [11] 2837-45 (1991).

Curtin W.A., “In-situ Fiber Strengths in Ceramic Matrix Composites from Fracture Mirrors,” *J. Am. Ceram. Soc.* **77** [4] 1075-78 (1994).

Curtin W.A., Ahn B.K. and Takeda N., “Modeling Brittle and Tough Stress-Strain Behavior in Unidirectional Ceramic Matrix Composites,” *Acta mater.* **46** [10] 3409-3420 (1998).

Davidge R.W., McLaren J.R. and Tappin G., ”Strength-probability-time (SPT) relationships in ceramics,” *Journal of Materials Science* **8** 1699-1705 (1973).

Forio P., Lavaire F. and Lamon J., “Delayed Failure at Intermediate Temperatures (600 – 700 °C) in Air in Silicon Carbide Multifilament Tows,” *J. Am. Ceram. Soc.* **87** [5] 888-893 (2004).

Gauthier W., Pailler F., Lamon J. and Pailler R., “Oxidation of Silicon Carbide Fibers During Static Fatigue in Air at Intermediate Temperatures,” *J. Am. Ceram. Soc.* **92** [9] 2067-2073 (2009).

Gauthier W. and Lamon J., “Delayed Failure of Hi-Nicalon and Hi-Nicalon S Multifilament Tows and Single Filaments at Intermediate Temperatures (500 °C – 800 °C),” *J. Am. Ceram. Soc.* **92** [3] 702-709 (2009).

Glime W.H. and Cawley J.D., “Stress Concentration Due to Fiber-Matrix Fusion in Ceramic-Matrix Composites,” *J. Am. Ceram. Soc.* **81** [10] 2597-604 (1998).

Hay R.S., “Growth Stress in SiO_2 During Oxidation of SiC Fibers,” *J. Appl. Phys.* **111** (2012).

List of References (continued)

Heredia F.E., McNulty J.C., Zok F.W. and Evans A.G., “Oxidation Embrittlement Probe for Ceramic Matrix Composites,” *J. Am. Ceram. Soc.* **78** [8] 2097-2100 (1995).

Kimber A.C. and Keer J.G., “On the Theoretical Average Crack Spacing in Brittle Matrix Composites Containing Continuous Aligned Fibers,” *Journal of Materials Science Letters*, **1** 353-354 (1982).

Lara-Curzio E., “Stress-Rupture of Nicalon/SiC Continuous Fiber Ceramic Composites in Air at 950 C,” *J. Am. Ceram. Soc.*, **80** [12] 3268-72 (1997).

Marshall D.B. and Evans A.G., “Failure Mechanisms in Ceramic Fiber/Ceramic Matrix Composites,” *J. Amer. Ceram. Soc.*, **68** [5] 225-31 (1985).

Morscher G.N., “Tensile Stress Rupture of SiCf/SiCm Minicomposites with Carbon and Boron Nitride Interphases at Elevated Temperatures in Air,” *J. Am. Ceram. Soc*. **80** [8] 2029-42 (1997).

Morscher G., Hurst J. and Brewer D., “Intermediate-Temperature Stress Rupture of a Woven Hi-Nicalon, BN-Interphase, SiC-Matrix Composite in Air,” *J. Am. Ceram. Soc.* 83 (6) 1441-49 (2000).

Morscher G.N. and Cawley J.D., “Intermediate temperature strength degradation in SiC/SiC composites,” *Journal of the European Ceramic Society* 22 (2002) 2777-2787.

Thouless M.D. and Evans A.G., “Effects of Pull-out on the Mechanical Properties of Ceramic-Matrix Composites,” *Acta metall*. **36** [3] 517-522 (1988).

Xu W., Zok F. and McMeeking R., “Model of Oxidation-Induced Fiber Fracture in SiC/SiC Composites,” *J. Am. Ceram. Soc.* 1-8 (2014).

Acknowledgements

This work was funded by the Transformational Tools and Technologies Project and the Transformative Aeronautics Concepts Program under NASA's Aeronautics Research Mission Directorate.